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The interaction between electrons and phonons in a thin carbon film is investigated. Sound wave propagation in a thin carbon film has a small sound velocity and small damping. The scattering of electrons by phonons associated with the Rayleigh wave is found to be responsible for the anomalous temperature dependence of the resistivity and for the negative magnetoresistance of pregraphitic carbons at low temperatures. (Keywords)

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ELECTRICAL CONDUCTION IN THIN FILM CARBONS[1]

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Electron-phonon interaction in a thin carbon film is investigated. Sound wave propagation in a thin carbon film has a small sound velocity and small damping. Electron-phonon scattering associated with the wave is responsible for the anomalous temperature dependence of the resistivity and for the negative magnetoresistance of pregraphitic carbons at low temperatures.

Lattice Vibrations in the Long Wavelength Approximation

We briefly describe the theory of the lattice vibrations in the long wave-length approximation. The equations of motion for lattice vibrations are given by:

$$\frac{\partial^2 u_x}{\partial t^2} = v_\ell^2 \frac{\partial^2 u_x}{\partial x^2} + v_t^2 \frac{\partial^2 u_x}{\partial y^2} + v_\ell^2 \left(\frac{1+\sigma}{2}\right)\frac{\partial^2 u_y}{\partial x \partial y} + \zeta\left[\frac{\partial^2 u_x}{\partial z^2} + \frac{\partial^2 u_z}{\partial z \partial x}\right],$$

$$\frac{\partial^2 u_z}{\partial t^2} = \zeta\left[\frac{\partial^2 u_z}{\partial x^2} + \frac{\partial^2 u_z}{\partial y^2}\right] + v_z^2\frac{\partial^2 u_z}{\partial z^2} + \zeta\left[\frac{\partial^2 u_x}{\partial z \partial x} + \frac{\partial^2 u_y}{\partial y \partial z}\right],$$

(1)

where $\vec{u} = (u_x, u_y, u_z)$ is a displacement vector. ζ is the elastic compliance, and v_l and v_t are the longitudinal and transverse velocities of sound associated with the in-plane vibrations, while v_z corresponds to the sound velocity for the out-of-plane vibrations. The equation for $\partial^2 u_y/\partial t^2$ is obtained by interchanging $x \leftrightarrow y$ in the equation for $\partial^2 u_x/\partial t^2$. The magnitude of other quantities in Eq. 1 are given by [1]

$$v_\ell = (C_{11}/\rho)^{\frac{1}{2}} = 2.10 \times 10^6 \text{ cm/sec},$$
$$v_t = [(C_{11} - C_{12})/2\rho]^{\frac{1}{2}} = 1.23 \times 10^6 \text{ cm/sec}, \quad (2)$$
$$v_z = (C_{33}/\rho)^{\frac{1}{2}} = 3.92 \times 10^5 \text{ cm/sec},$$

where the density $\rho = 2.26\text{g/cm}^3$, and the Poisson ratio $\sigma = C_{12}/C_{11}$. The elastic compliance ζ is related to the elastic constant C_{44} by $\zeta = C_{44}/\rho$. Both C_{44} and ζ are thus related to the shearing force and are very sensitive to crystal perfection, especially to the density of stacking faults. The magnitude of C_{44} thus ranges from 4.5×10^{10}dynes/cm² for well-graphitized crystals [1,2] to 7.0×10^9dynes/cm² for pregraphitic carbons with a high density of stacking faults [3,4].

In the expression for u_z in Eq. 1 the term related to the bonding between the honeycomb planar networks is neglected, since this term is negligible in the long wave-length limit [5]. The sound velocity v_R of the Rayleigh wave in thin film carbons is approximately given by $\sqrt{\zeta}$.

Electron–Rayleigh Wave Interaction in Thin Carbon Films

If the sample thickness d is small (< 100 Å), a Rayleigh wave with small sound velocity ($\sim 10^4$ cm/sec) can propagate in the film without damping. The Rayleigh wave is polarized along the c-axis. Carriers are strongly scattered by the phonons associated with this Rayleigh wave even at $T \leq 1$K, since typical phonon energies interacting with carriers are at most 1K. Though the carrier relaxation rate relative to the interaction with the Rayleigh wave phonons is one order of magnitude smaller than that for impurity scattering, the electron–Rayleigh wave interaction plays an important role in the anomalous temperature dependence of the resistivity and in the negative magnetoresistance of disordered carbons at low temperature [6].

Pregraphitic carbons heat treated at low temperature have a turbostratic structure and the correlation length along the c-axis is small. Accordingly, it is reasonable to assume that the sample is composed of an assembly of many thin films which are weakly coupled to each other elastically.

To solve Eq. 1 for the case of a thin film, the following boundary conditions are imposed at $z = 0$ and $z = -d$, where d is the film thickness along the c-axis. Strain free and stress free boundary conditions give rise to the same equations:

$$\frac{\partial u_x}{\partial z} + \frac{\partial u_z}{\partial x} = \frac{\partial u_y}{\partial z} + \frac{\partial u_z}{\partial y} = \frac{\partial u_z}{\partial z} = 0, \quad (3)$$

at $z = 0$ and $z = -d$. A damped plane wave solution of Eq. 1 is written in the form:

$$\vec{u}(r) = \vec{U}e^{\kappa z}e^{i(qx - \omega t)} \quad (4)$$

for $\vec{q}$ along the x-axis. The general solution $\vec{u}(r) \propto e^{i(\vec{q}\cdot\vec{r}-\omega t)}$ can be obtained from Eq. 4 by rotating the coordinate axis in the xy-plane. Inserting Eq. 4 into Eq. 1, yields $u_y = 0$ and equations for the u_x and u_z components:

$$(\omega^2 - v_\ell^2 q^2 + \zeta\kappa^2)U_x + i\zeta q\kappa U_z = 0$$
$$i\zeta q\kappa U_x + (\omega^2 - \zeta q^2 + v_z^2\kappa^2)U_z = 0. \quad (5)$$

Equation (5) is solved to obtain κ^2 as a function of ω. Two positive roots κ_1^2 and κ_2^2 exist, if the Rayleigh wave velocity $v_R = \omega/q$ satisfies the condition:

$$v_R^2 < \zeta = C_{44}/\rho. \quad (6)$$

One root corresponds to a rapidly damped wave and the other to a weakly damped wave. From these roots, an explicit expression for the displacement vector $\vec{u} = (u_x, 0, u_z)$ has been found. By introducing the phonon operators b_q^+ and b_q, the energy of the Rayleigh wave is quantized according to

$$2 \int d\vec{r} \ \frac{\rho}{2} \Big\{ |\ \frac{\partial \vec{u}_a}{\partial t}\ |^2 + |\ \frac{\partial \vec{u}_z}{\partial t}\ |^2 \Big\} = \sum_q \hbar\omega_q (b_q^+ b_q + \frac{1}{2}), \quad (7)$$

where $\vec{u}_a = (u_x, u_y, 0)$. From these equations, explicit expressions for $\vec{u}_a$ and $\vec{u}_z$ are obtained [6].

The solutions show that the displacements follow a weakly damped Rayleigh wave polarized along the c–axis where the in–plane displacement $\vec{u}_a$ is two orders of magnitude smaller than $\vec{u}_z$. The following expression for the k–dependent relaxation rate due to the scattering by the Rayleigh wave phonons is obtained [6]:

$$1/\tau_R(E_k) \simeq \frac{2\pi k_B T D^2}{\hbar \rho v_R^2 d^2 \Omega} \sum_{\vec{k}'} \frac{1}{q^2}\Big(1 - \frac{k'_z}{k_z}\Big) \delta(E_k - E_{k'}), \quad (8)$$

where $\vec{q} = \vec{k}'_a - \vec{k}_a$, and $\vec{k}_a = (k_x, k_y, 0)$. Here D denotes the electron–phonon coupling constant associated with the out–of–plane vibrations, and in bulk graphite $D \sim 3.7\text{eV}[7]$. In deriving Eq. 8, the introduction of a high temperature approximation for phonons $N_q \sim N_q + 1 \sim k_B T/\hbar\omega_q$ is employed, since $\hbar\omega_q/k_B = \hbar v_R q/k_B \simeq 4 \times 10^7 q < 1\text{K}$ for $q \sim 10^6\text{cm}^{-1}$. From Eq. 8, we then obtain a temperature dependence for the relaxation rate

$$\frac{1}{\tau_R} \simeq \frac{k_B T}{4\pi^2 \hbar^2 \rho c_0} \Big(\frac{1}{v_F\ q_{min}}\Big)\Big(\frac{D}{v_R d}\Big)^2, \quad (9)$$
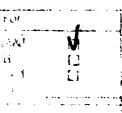
where $q_{min} = 2\pi/\hat{L}_a$ and $\hat{L}_a$ denotes the dimension of the thin film in the basal–plane. In evaluating Eq. 9, the following values of the parameters are employed: $v_R = 4.5 \times 10^4$ cm/sec, $d = 70\text{Å}$, $L_a = 200\text{Å}$, and $v_F = 5 \times 10^7$ cm/sec, yielding a value of $1/\tau_R \sim 3 \times 10^{11}\text{T/secK}$. The large magnitude of $1/\tau_R$ compared with other processes in thin carbon films shows that the electron–Rayleigh wave interaction is responsible for the anomalous temperature dependent negative magnetoresistance of pre-graphitic carbons at low temperatures[6], and for the unusual temperature dependence of the carbon films with low heat treatment temperature $T_{HT} \simeq 1300°\text{C}$ [8].

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